

FORM PTO-1390 (REV. 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER 0020-4961P
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			U.S. APPLICATION NO. (If known, see 37 CFR 1.5) 10/069342
INTERNATIONAL APPLICATION NO. PCT/JP00/05673	INTERNATIONAL FILING DATE August 24, 2000	PRIORITY DATE CLAIMED August 25, 1999	
TITLE OF INVENTION LAMINATE COMPRISING FLUORINE-CONTAINING POLYMER			
APPLICANT(S) FOR DO/EO/US HIGUCHI, Tatsuya; OZAKI, Hidenori; KITAHARA, Takahiro; ISHIWARI, Kazuo			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<p>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).</p> <p>4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p> a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</p> <p> b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau. WO 01/14141</p> <p> c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</p> <p>6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).</p> <p> a. <input checked="" type="checkbox"/> is transmitted herewith.</p> <p> b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4)</p> <p>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p> a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</p> <p> b. <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p> c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p> d. <input checked="" type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p>			
Items 11. to 20. below concern document(s) or information included:			
<p>11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98, Form PTO-1449(s), and International Search Report (PCT/ISA/210) with 0 cited document(s).</p> <p>12. <input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment.</p> <p>14. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>15. <input type="checkbox"/> A substitute specification.</p> <p>16. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821-1.825.</p> <p>18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</p> <p>19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).</p> <p>20. <input checked="" type="checkbox"/> Other items or information:</p> <p> 1.) PCT/IB/304 and PCT/IB/308</p> <p> 2.) Zero (0) sheets of Formal Drawings</p>			

(see 37 CFR 1.5)

1069342

INTERNATIONAL APPLICATION NO

PCT/JP00/05673

ATTORNEY'S DOCKET NUMBER

0020-4961P

The following fees are submitted:

CALCULATIONS PTO USE ONLY

NATIONAL FEE (37 CFR 1.492(a)(1)-(5):

International preliminary examination fee (37 CFR 1.482)
 or international search fee (37 CFR 1.445(a)(2)) paid to USPTO
 and International Search Report not prepared by the EPO or JPO \$1,040.00

International preliminary examination fee (37 CFR 1.482) not paid to
 USPTO but International Search Report prepared by the EPO or JPO \$890.00

International preliminary examination fee (37 CFR 1.482) not paid to USPTO
 but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$740.00

International preliminary examination fee (37 CFR 1.482) paid to USPTO
 but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$710.00

International preliminary examination fee (37 CFR 1.482) paid to USPTO
 and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00

ENTER APPROPRIATE BASIC FEE AMOUNT =

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30
 months from the earliest claimed priority date (37 CFR 1.492(e)).

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total Claims	15 - 20 =	0	X \$18.00	\$	0
Independent Claims	1 - 3 =	0	X \$84.00	\$	0
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			None	+ \$280.00	\$ 0

TOTAL OF ABOVE CALCULATIONS = \$ 890.00

☐ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are
 reduced by 1/2.

SUBTOTAL = \$ 890.00

Processing fee of \$130.00 for furnishing the English translation later than ☐ 20 ☐ 30
 months from the earliest claimed priority date (37 CFR 1.492(f)).

TOTAL NATIONAL FEE = \$ 890.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be
 accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +

TOTAL FEES ENCLOSED = \$ 930.00

Amount to be:
 refunded \$
 charged \$

- ☒ A check in the amount of \$ **930.00** to cover the above fees is enclosed.
☐ Please charge my Deposit Account. No. _____ in the amount of \$ _____ to cover the above fees.
 A duplicate copy of this sheet is enclosed.
☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
 overpayment to Deposit Account No. **02-2448**.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR
 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

Send all correspondence to:

Birch, Stewart, Kolasch & Birch, LLP or Customer No. 2292
 P.O. Box 747
 Falls Church, VA 22040-0747
 (703) 205-8000

Date: February 25, 2002

By Andrew D. Meikle
 (Andrew D. Meikle, #32,868)

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicant: HIGUCHI, Tatsuya et al.
Int'l. Appl. No.: PCT/JP00/05673
Appl. No.: New Group:
Filed: February 25, 2002 Examiner:
For: LAMINATE COMPRISING FLUORINE-
CONTAINING POLYMER

PRELIMINARY AMENDMENT

BOX PATENT APPLICATION

Assistant Commissioner for Patents
Washington, DC 20231

February 25, 2002

Sir:

The following Preliminary Amendments and Remarks are respectfully submitted in connection with the above-identified application.

AMENDMENTS

IN THE SPECIFICATION:

Please amend the specification as follows:

Before line 1, insert --This application is the national phase under 35 U.S.C. § 371 of PCT International Application No. PCT/JP00/05673 which has an International filing date of August 24, 2000, which designated the United States of America.--

IN THE CLAIMS:

Please amend the claims as follows:

3. (Amended) The laminate according to claim 1, wherein said tetrafluoroethylene copolymer comprises 40 to 81 % by mole of tetrafluoroethylene and 60 to 19 % by mole of other monomer.

4. (Amended) The laminate according to claim 1, wherein said tetrafluoroethylene copolymer has a melt flow rate of 1.0 to 100 g/10 minutes.

5. (Amended) The laminate according to claim 1, wherein said tetrafluoroethylene copolymer has a melt flow rate of 1.0 to 50 g/10 minutes.

6. (Amended) The laminate according to claim 1, wherein said tetrafluoroethylene copolymer has a critical shear rate of 10 to 10^3 sec^{-1} at 230°C.

7. (Amended) The laminate according to claim 1, wherein said tetrafluoroethylene copolymer comprises tetrafluoroethylene, ethylene, a fluorovinyl compound of the formula (I):



wherein Rf is a fluoroalkyl group having 2 to 10 carbon atoms, and hexafluoropropylene, in which a molar ratio of

tetrafluoroethylene to ethylene is from 40:60 to 90:10, the content of said fluorovinyl compound is from 0 to 10 % by mole (based on the whole copolymer) and the content of hexane is from 0 to 30 % by mole (based on the whole copolymer).

8. (Amended) The laminate according to claim 1, wherein said ethylene-vinyl acetate copolymer has a vinyl acetate content X (% by mole) and a saponification degree Y (%) of a methyl ester satisfy the following relationship:

$$X \times Y/100 \geq 10.0.$$

9. (Amended) The laminate according to claim 1, wherein said ethylene-vinyl acetate copolymer has a melt flow rate (200°C, 5 kg-load) of 0.5 to 100 g/10 minutes.

10. (Amended) A tubular member comprising a laminate according to claim 1.

12. (Amended) A container comprising a laminate according to claim 1.

14. (Amended) A film comprising a laminate according to claim 1.

15. (Amended) A sheet comprising a laminate according to claim 1.

REMARKS

The specification has been amended to provide a cross-reference to the previously filed International Application.

The amendment to the claims is merely to delete the multiple dependencies and to place the application into better form for examination. Entry of the above amendments is earnestly solicited. An early and favorable first action on the merits is earnestly solicited.

Attached hereto is a marked-up version of the changes made to the application by this Amendment.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17; particularly, extension of time fees.

Respectfully submitted,

BIRCH, STEWART, KOLASCH & BIRCH, LLP

By 
Andrew D. Meikle, #32,868

ADM/cqc
0020-4961P

P.O. Box 747
Falls Church, VA 22040-0747
(703) 205-8000

Attachment: VERSION WITH MARKINGS TO SHOW CHANGES MADE

(Rev. 02/21/02)

VERSION WITH MARKINGS TO SHOW CHANGES MADE

The claims have been amended as follows:

3. (Amended) The laminate according to claim 1 or 2, wherein said tetrafluoroethylene copolymer comprises 40 to 81 % by mole of tetrafluoroethylene and 60 to 19 % by mole of other monomer.

4. (Amended) The laminate according to [any one of claims 1 to 3]claim 1, wherein said tetrafluoroethylene copolymer has a melt flow rate of 1.0 to 100 g/10 minutes.

5. (Amended) The laminate according to [any one of claims 1 to 3]claim 1, wherein said tetrafluoroethylene copolymer has a melt flow rate of 1.0 to 50 g/10 minutes.

6. (Amended) The laminate according to [any one of claims 1 to 5]claim 1, wherein said tetrafluoroethylene copolymer has a critical shear rate of 10 to 10^3 sec^{-1} at 230°C.

7. (Amended) The laminate according to [any one of claims 1 to 6]claim 1, wherein said tetrafluoroethylene copolymer comprises tetrafluoroethylene, ethylene, a fluorovinyl compound of the formula (I):



wherein Rf is a fluoroalkyl group having 2 to 10 carbon atoms,

and hexafluoropropylene, in which a molar ratio of tetrafluoroethylene to ethylene is from 40:60 to 90:10, the content of said fluorovinyl compound is from 0 to 10 % by mole (based on the whole copolymer) and the content of hexane is from 0 to 30 % by mole (based on the whole copolymer).

8. (Amended) The laminate according to [any one of claims 1 to 7]claim 1, wherein said ethylene-vinyl acetate copolymer has a vinyl acetate content X (% by mole) and a saponification degree Y (%) of a methyl ester satisfy the following relationship:

$$X \times Y/100 \geq 10.0.$$

9. (Amended) The laminate according to [any one of claims 1 to 8]claim 1, wherein said ethylene-vinyl acetate copolymer has a melt flow rate (200°C, 5 kg-load) of 0.5 to 100 g/10 minutes.

10. (Amended) A tubular member comprising a laminate according to [any one of claims 1 to 9]claim 1.

12. (Amended) A container comprising a laminate according to [any one of claims 1 to 9]claim 1.

14. (Amended) A film comprising a laminate according to [any one of claims 1 to 9]claim 1.

15. (Amended) A sheet comprising a laminate according to
[any one of claims 1 to 9]claim 1.

(Rev. 11/13/01)

DESCRIPTION

LAMINATE COMPRISING FLUORINE-CONTAINING POLYMER

5 FIELD OF THE INVENTION

The present invention relates to a laminate comprising a fluorine-containing polymer. In particular, the present invention relates to a laminate comprising a layer of a tetrafluoroethylene copolymer having a carbonate group and a layer
10 of a polyolefine resin.

BACKGROUND ART

Fluorine-containing polymers are used in various applications since they have excellent heat resistance, chemical resistance, surface properties (e.g. low abrasion properties,
15 etc.) and electrical insulating properties. However, the fluorine-containing polymers have generally insufficient mechanical strength or dimensional stability, or they are expensive.

Thus, various attempts such as the adhesion or lamination
20 of the fluorine-containing polymer to other organic material or an inorganic material have been made to make the best use of advantages of the fluorine-containing polymers while minimizing their disadvantages.

However, the fluorine-containing polymers inherently have
25 a low adhesion force so that it is difficult to adhere them directly to other material. Even if fusion bonding is tried, the adhesion strength is insufficient, or when a certain level of adhesion strength is attained, the adhesion force tends to vary with a kind

of a substrate. Thus, the reliability of the adhesion is often insufficient.

To adhere the fluorine-containing polymers to other material, the following methods are mainly discussed:

5 1. Physically roughening the surface of a substrate with sand bluster treatment, etc.;

 2. Surface treatment of a fluorine-containing polymer by sodium etching, plasma treatment, photochemical treatment, etc.;

 3. Adhesion with an adhesive.

10 The methods 1 and 2 require the treatment process and the treatment steps are complicated, and thus their productivity is low. In addition, in the methods 1 and 2, the kinds and shapes of the substrates are limited. Anyhow, the adhesion force achieved by the methods 1 and 2 is low, and the appearance of the resulting
15 laminate tends to have some problems (e.g. coloring, flaws, etc.).

 Various adhesives to be used in the method 3 are examined. Conventional hydrocarbon base adhesives have insufficient adhesion properties, and their own heat resistance is insufficient. Therefore, such adhesives cannot generally withstand adhering
20 conditions for the fluorine-containing polymers which require molding or processing at high temperature, and they suffer from delamination or coloration due to decomposition. The laminates using such adhesives cannot maintain the adhesion force due to the change of temperature and environment and lack reliability
25 since their adhesive layers have insufficient heat resistance, chemical resistance or water resistance.

 Alternatively, adhesion with an adhesive or an adhesive composition comprising a fluorine-containing polymer having a

functional group is studied.

Prior arts disclose, as an adhesive, the use a fluorine-containing polymer to which a hydrocarbon monomer having a carboxyl group, a carboxylic anhydride group, an epoxy group or a hydrolyzable silyl group (e.g. maleic anhydride, vinyltrimethoxysilane, etc.) is grafted (e.g. JP-A-7-18035, JP-A-7-25952, JP-A-7-25954, JP-A-7-173230, JP-A-7-173446, JP-A-7-173447, etc.), or the use of a cured material of an adhesive composition containing a fluorine-containing copolymer which comprises a hydrocarbon monomer having a functional group such as hydroxyalkyl vinyl ether copolymerized with tetrafluoroethylene, chlorotrifluoroethylene, etc., and a isocyanate curing agent as an adhesive for adhering polyvinyl chloride to ETFE (ethylene-tetrafluoroethylene copolymer) which has been treated with corona discharge (e.g. JP-A-7-228848).

The adhesive or adhesive composition comprising the fluorine-containing polymer to which the functional hydrocarbon monomer is grafted or copolymerized has insufficient heat resistance. Thus, it is decomposed and foamed so that the adhesion strength decreases, or the layers are delaminated or colored, when it is processed together with the fluororesin or used at high temperature. The adhesive composition disclosed in JP-A-7-228848 requires the corona discharge treatment of the fluororesin.

Furthermore, it is reported to use a fluorine-containing polymer having a functional group, which is prepared by copolymerizing a perfluorovinyl ether having a carboxylic acid group or its derivative with a fluorine-containing monomer, as an adhesive or an adhesive composition. US Patent No. 4,916,020

discloses a laminate using a fluorine-containing polymer having a functional group which is introduced in the polymer by copolymerizing a perfluorovinyl ether having a carboxylic acid group or its derivative with tetrafluoroethylene, etc.

5 In this laminate, the above-described fluorine-containing polymer having a carboxylic acid group, etc. is laminated on other substrate such as a metal substrate through an adhesive resin such as an epoxy resin, a urethane resin, etc. Thus, the epoxy resin or the urethane resin has problems in its heat resistance, chemical
10 resistance or solvent resistance when the laminate is used.

WO98/58973 (published on December 30, 1998) discloses a laminate comprising a layer of a tetrafluoroethylene copolymer having a terminal carbonate group, a layer of other material such as a thermoplastic resin, and an intermediate layer between them.
15 One example described in this WO publication is a laminate comprising an intermediate layer of an epoxy group-containing polyethylene when polyethylene is used as the other material.

When the layer of the tetrafluoroethylene copolymer having a terminal carbonate group and the polyethylene layer are adhered
20 with the epoxy group-containing polyethylene, the initial adhesion force is high, but the adhesion force tends to decrease with time. In addition, the epoxy group-containing polyethylene itself has low resistance to fuel and is dissolved in the fuel. Thus, this laminate cannot be used to fabricate a tube or a tank
25 for storing the fuel.

DISCLOSURE OF THE INVENTION

One object of the present invention is to provide a laminate which comprises a layer of a tetrafluoroethylene copolymer having

a carbonate group and a layer of a polyolefine resin strongly adhered each other, the adhesion strength of which does not decrease with time, and which has high resistance to fuel.

According to the present invention, the above object is
5 achieved by a laminate comprising

(1) a layer of a tetrafluoroethylene copolymer comprising 30 to 81 % by mole of tetrafluoroethylene and 70 to 19 % by mole of at least one other monomer and having a carbonate group in a polymer chain or at a polymer chain terminal, which has a melt
10 flow rate of 0.1 to 100 g/10 minutes (230°C, 5 kg-load) and a melting point of 90 to 230°C,

(2) a layer of an ethylene-vinyl acetate copolymer formed on one surface of the layer (1), which satisfies the following relationship:

15
$$X \times Y/100 \geq 7.0$$

wherein X is a vinyl acetate content (% by mole) and Y is a saponification degree of a methyl ester (%), and

(3) a layer of a polyolefin resin formed on the layer (2).

The carbonate group contained in the tetrafluoroethylene
20 copolymer to be used in the present invention is a group having a bond of the formula: $-\text{OC}(=\text{O})\text{O}-$, and a specific example of the carbonate group is a group of the formula: $-\text{OC}(=\text{O})\text{O}-\text{R}$ wherein R is a hydrogen atom, an organic group (e.g. a C_1 - C_{20} alkyl group, a C_2 - C_{20} alkyl group having an ether linkage, etc.), or an element
25 of the I, II or VII Group. Preferable examples of the carbonate group include $-\text{OC}(=\text{O})\text{O}-\text{CH}_3$, $-\text{OC}(=\text{O})\text{O}-\text{C}_3\text{H}_7$, $-\text{OC}(=\text{O})\text{O}-\text{C}_8\text{H}_{17}$, $-\text{OC}(=\text{O})\text{O}-\text{CH}_2\text{CH}_2\text{OCH}_2\text{CH}_3$, and the like.

The tetrafluoroethylene copolymer having a carbonate group

at a polymer chain terminal means a tetrafluoroethylene polymer having a functional group comprising a carbonate group on at least one terminal of the polymer chain. The tetrafluoroethylene copolymer having a carbonate group in a polymer chain means a
5 copolymer prepared by copolymerizing an ethylenically unsaturated monomer having a carbonate group and tetrafluoroethylene. One polymer chain may have the carbonate groups in the polymer chain and also at the polymer chain terminal.

Among these copolymers, the tetrafluoroethylene copolymer
10 having a carbonate group at a polymer chain terminal is preferable since it does not remarkably deteriorate heat resistance, mechanical properties and chemical resistance, or it is advantageous from the viewpoint of productivity and cost.

A polymer having no carbonate group may be compounded in
15 the tetrafluoroethylene copolymer having a carbonate group.

According to the present invention, it is necessary for the tetrafluoroethylene copolymer to be fusion bonded to the polyolefin layer as an opposite material for lamination at a temperature at which polyolefin is not thermally decomposed, and
20 has a viscosity so that the copolymer can sufficiently flow at such a temperature. A preferred range of the fusion bonding temperature of polyolefin is from about 200 to 280°C. Thus, the tetrafluoroethylene copolymer used in the present invention should be melt and have a melt viscosity so that it can flow in
25 this temperature range.

When it is desired to lower the permeability of a fuel, the tetrafluoroethylene copolymer having a high melting point, that is, a melt flow rate of 0.1 to 100 g/10 minutes (230°C, 5 kg-

load) and a melting point of 90 to 230°C is preferably used. When the other material has low heat resistance, the tetrafluoroethylene copolymer having a low melting point, that is, a melt flow rate of 0.1 to 100 g/10 minutes (200°C, 5 kg-load) and a melting point of 90 to 200°C is preferably used.

In the present invention, the melt flow rate (MFR) of the tetrafluoroethylene copolymer is preferably from 1.0 to 100 g/10 minutes, more preferably from 1.0 to 50 g/10 minutes.

Herein, the melt flow rate is measured at 200°C or 230°C under a load of 5 kg with an orifice diameter of 2 mm and a land length of 8 mm.

In addition, the tetrafluoroethylene copolymer used in the present invention has a critical shear rate of 10 to 10^3 sec^{-1} at 230°C, and can be molded at a lower temperature than conventional fluororesins. Because of such a critical shear rate, the tetrafluoroethylene copolymer used in the present invention can solely be molded to form a molded article in the form of a film or a tube and also coextrusion molded with the polyolefin resin.

In one preferred embodiment, the tetrafluoroethylene copolymer used in the present invention has a monomer composition comprising 40 to 81 % by mole of tetrafluoroethylene (TFE) and 60 to 19 % by mole of other monomer, in particular, 40 to 81 % by mole of TFE, 6 to 43 % by mole of ethylene (Et) and 0 to 30 % by mole of hexafluoropropylene (HFP).

More preferably, the monomer composition comprises 40 to 60 % by mole of TFE, 24 to 43 % by mole of Et and 5 to 30 % by mole of HFP, since a polymerization decreases as the content of TFE increases.

The melting point of the tetrafluoroethylene copolymer used in the present invention continuously decreases and its crystallinity decreases so that its transparency increases, as the content of HFP increases.

5 In another preferred embodiment, in addition to TFE, Et and HFP, the tetrafluoroethylene copolymer used in the present invention may contain, as a modifier monomer, a fluorovinyl compound of the formula (I):



10 wherein Rf is a fluoroalkyl group having 2 to 10 carbon atoms.

When the number of carbon atoms of Rf is less than 2, the modification of the tetrafluoroethylene copolymer (for example, the suppression of cracking of molded articles during molding of the copolymer or in use) may be insufficient. When the number
15 of carbon atoms of Rf exceeds 10, the compound (I) may be disadvantageous from the viewpoint of polymerization reactivity. In particular, Rf having 3 to 5 carbon atom is preferable.

From the viewpoint of the heat resistance of the copolymer obtained, the Rf group is most preferably a perfluoroalkyl group,
20 or a ω -hydro- or ω -chloroperfluoroalkyl group.

Among those fluorovinyl compounds, a fluorovinyl compound of the formula (II):



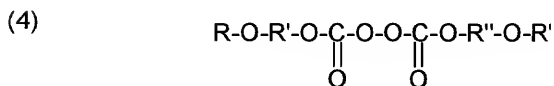
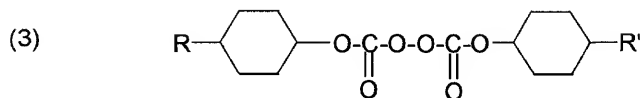
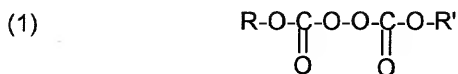
wherein n is a number of 2 to 10, in particular, a number of 3
25 to 5

is preferable from the viewpoint of copolymerizability, costs for the preparation of the copolymer, and the properties of the copolymer obtained.

When the fluorovinyl compound (I) is used as a modifier monomer, a monomer composition is such that a molar ratio of tetrafluoroethylene to ethylene is from 40:60 to 90:10, the content of the fluorovinyl compound is from 0 to 10 % by mole (based on the whole copolymer) and the content of hexane is from 0 to 30 % by mole (based on the whole copolymer).

The tetrafluoroethylene copolymer of the present invention can be prepared by a polymerization process which is used to prepare ETFE.

To introduce the carbonate group at the chain terminal, a peroxy carbonate is used as a polymerization initiator in the preparation of the tetrafluoroethylene copolymer of the present invention. Examples of the peroxy carbonate include the following compounds (1) to (4):



wherein R and R' represent independently each other a monovalent

saturated straight or branched hydrocarbon group having 1 to 15 carbon atoms or a monovalent saturated straight or branched hydrocarbon having 1 to 15 carbon atoms and an alkoxyl group at the end of the molecule, and R" is a divalent saturated straight or branched hydrocarbon group having 1 to 15 carbon atoms or a divalent saturated straight or branched hydrocarbon having 1 to 15 carbon atoms and an alkoxyl group at the end of the molecule.

In particular, diisopropyl peroxydicarbonate, di-n-propyl peroxydicarbonate, tert.-butylperoxyisopropyl carbonate, bis(4-tert.-butylcyclohexyl) peroxydicarbonate, di-2-ethylhexyl peroxydicarbonate, etc. are preferable.

Industrially, the polymerization is preferably carried out by suspension polymerization in an aqueous medium using a fluorine-containing solvent and a peroxydicarbonate as a polymerization initiator, although other polymerization methods such as solution polymerization, bulk polymerization, etc. may be employed.

Examples of the fluorine-containing solvent include hydrochlorofluoroalkanes (e.g. CH_3CClF_2 , $\text{CH}_3\text{CCl}_2\text{F}$, $\text{CF}_3\text{CF}_2\text{CCl}_2\text{H}$, $\text{CF}_2\text{ClCF}_2\text{CFHCl}$, etc.), chlorofluoroalkanes (e.g. $\text{CF}_2\text{ClCFClCF}_2\text{CF}_3$, $\text{CF}_3\text{CFClCFClCF}_3$, etc.), and perfluoroalkanes (e.g. perfluorocyclobutane, $\text{CF}_3\text{CF}_2\text{CF}_2\text{CF}_3$, $\text{CF}_3\text{CF}_2\text{CF}_2\text{CF}_2\text{CF}_3$, $\text{CF}_3\text{CF}_2\text{CF}_2\text{CF}_2\text{CF}_2\text{CF}_3$, etc.). Among them, the perfluoroalkanes are preferable.

The amount of the solvent used is preferably from 10 to 100 % by weight based on water from the viewpoint of suspending properties and costs.

The polymerization temperature is not limited, and is

preferably from 0 to 100°C.

The polymerization pressure may be determined according to other polymerization conditions such as the kind, amount and vapor pressure of the solvent used, the polymerization temperature, etc.

5 Usually, the polymerization pressure may be from 0 to 50 kgf/cm²G.

In the preparation of the tetrafluoroethylene copolymer of the present invention, a conventional chain transfer agent, for example, a hydrocarbon such as isopentane, n-pentane, n-hexane, cyclohexane; an alcohol such as methanol, ethanol, etc.; and a
10 halohydrocarbon such as carbon tetrachloride, chloroform, methylene chloride, methyl chloride, etc., can be used to adjust the molecular weight.

As the ethylene-vinyl acetate copolymers, those having various contents of vinyl acetate and various saponification
15 degrees are commercially available. Thus, from those ethylene-vinyl acetate copolymers, one satisfying the relationship of $X \times Y/100 \geq 7.0$ wherein X is a vinyl acetate content (% by mole) and Y is a saponification degree of a methyl ester(%)
20 can be selected.

For example, EVAL 101, which is available from KURARAY Co., Ltd., has a vinyl acetate content of 68 % and a saponification degree of 95 %. Thus, $X \times Y/100$ is 4.6. MELSEN H6051, which is available from TOSO CORPORATION, has a vinyl content of 11.2 %
25 and a saponification degree of 100 %. Thus, $X \times Y/100$ is 11.2. TECHNOLINK K200, which is available from TAOKA Chemical Co., Ltd., has a vinyl acetate content of 11.2 % and a saponification degree of 85 %. Thus, $X \times Y/100$ is 9.52. They are all ethylene-vinyl

acetate copolymers which can be used according to the present invention.

The melt flow rate (200°C, 5 kg-load) is from 0.5 to 100 g/10 minutes, preferably from 1.0 to 50g/10 minutes.

5 The viscosity of the ethylene-vinyl acetate copolymer is preferably close to that of the tetrafluoroethylene copolymer. When the difference of the viscosities of the two copolymers is large, the thickness of either the layer of the tetrafluoroethylene copolymer or the layer of the ethylene-vinyl acetate copolymer may become uneven. Therefore, the melt flow
10 rate of the ethylene-vinyl acetate copolymer is preferably in the above range.

As the polyolefin resin, preferably polyethylene, polystyrene, polypropylene, polybutene, ethylene-vinyl acetate
15 copolymer (EVA), ethylene-methyl acetate copolymer, ethylene-ethyl acrylate copolymer, ethylene-ethyl acrylate-maleic anhydride copolymer, and the like are used. In particular, polyethylene is preferable.

To increase the adhesion strength between the ethylene-vinyl acetate copolymer layer (2) and the polyolefin resin layer (3), a known adhesive olefin (e.g. maleic anhydride-modified polyethylene, maleic anhydride-modified polypropylene, etc.), maleic anhydride-modified polystyrene, and the like may be used as an adhesive.

25 Each of the layers of the laminate according to the present invention may contain an additive which is suitable for each polymer such as a reinforcing agent, a filler, a stabilizer, a UV ray absorber, a pigment, etc. insofar as the characteristics

of each layer are not deteriorated.

Such an additive can improve heat stability, surface hardness, abrasion resistance, antistatic properties, weather resistance, or other properties.

5 A method for producing the laminate of the present invention may be properly selected in accordance with the kind of the tetrafluoroethylene copolymer, the kinds of the materials used to form other layers, etc.

For example, the polymeric materials which form the
10 respective layers are multilayer coextruded to form all the layers at one time to produce a molded article in the form of a film, a sheet, a tube, a hose, a bottle, a tank, etc.

EXAMPLES

Example 1

15 Pellets of a fluorine-containing polymer (monomer composition (molar ratio: TFE/Et/HFP/H2P ($\text{CH}_2=\text{CF}(\text{CF}_2)_3\text{H}$) = 46.2/35.8/17.5/0.5; melting point: 158°C; MFR: 20 g/10 min. (230°C, 5 kg-load)) were charged in a mold having a diameter of 120 mm, and the mold is set in a press heated at 230°C, and the polymer
20 was melt pressed under a pressure of about 50 kgf/cm² to obtain a sheet having a thickness of 0.5 mm.

Separately, each of an ethylene-vinyl alcohol copolymer (EVOH) (EVAL F 101A available from KURARAY Co., Ltd.; vinyl acetate content X: 68 % by mole; saponification degree: 95 %; X x Y/100
25 = 64.6), maleic anhydride-modified polyethylene (ADOMER NF 528 available from MITSUI CHEMICALS, INC.) and high density polyethylene (BATEC-HD HJ560 available from Japan Polychem Corporation) was pressed at 230°C in the same manner as above to

obtain a sheet having a thickness of 0.5 mm.

Then, the four sheets obtained in the above steps were laminated in the order of the fluorine-containing polymer/ethylene-vinyl alcohol copolymer/maleic anhydride-
5 modified polyethylene/high density polyethylene with inserting a polyimide film as a spacer partly between the fluorine-containing polymer sheet and the ethylene-vinyl alcohol copolymer sheet, and the laminate was placed in a mold having a diameter of 120 mm. The mold was set in a press heated at 230°C and the
10 laminate was pressed for 10 seconds under a pressure of 10 kgf/cm² to fusion bond the sheets.

The polyimide film spacer was removed, and the unbonded parts were provided as grasping parts in an adhesion strength test. Then, a maximum peel strength (initial) and a fuel-resistant
15 strength were measured by a T-shape peeling test method with a Tensilon general tester (manufactured by ORIENTEC Co., Ltd.).

As a fuel-resistant strength, a maximum peel strength was used, which was measured after dipping the sample in a fuel (CM15: a mixture of isoocatane and toluene in a volume ratio of 50:50
20 containing 15 % by volume of methanol) at 60°C for 168 hours.

The results are shown in Table 1.

Examples 2-3 and Comparative Examples 1-4

A sheet was produced in the same manner as in Example 1 except that the layer structure was changed and the materials of each
25 layer were selected as described below. The results are shown in Table 1.

(Example 2)

Layer structure: TFE copolymer layer/ethylene-vinyl

acetate copolymer layer/polyolefin layer

TFE copolymer: the same as Example 1

Ethylene-vinyl acetate copolymer: EVOH (MELSEN H6051,
available from TOSO CORPORATION; acetate content X: 11.2 %;

5 saponification degree: 100 %; $X \times Y/100$: 11.2)

Polyolefin: low density polyethylene (PETROCENE 292
available from TOSO CORPORATION)

(Comparative Example 1)

Layer structure: TFE copolymer layer/epoxy group-
10 containing polyethylene layer/polyolefin layer

TFE copolymer: the same as Example 1

Epoxy group-containing polyethylene: epoxy-modified
polyethylene (LEX PERL RA3150 available from Nippon Polyolefins
Co., Ltd.)

15 Polyolefin: polyethylene (UBE POLYETHYLENE L519 available
from Ube Industries, Ltd.)

(Example 3)

Layer structure: the same as Example 1

The same as those used in Example 1 except that EVOH
20 (TECHNOLINK K200 available from TAOKA Chemical Co., Ltd.; vinyl
acetate content: 11.2 %; saponification degree: 85 %; $X \times Y/100$:
9.52) being used as an ethylene-vinyl acetate copolymer layer.

(Comparative Example 2)

Layer structure: the same as Example 1

25 The same as those used in Example 1 except that EVOH (MELSEN
H 6410M available from TOSO CORPORATION; vinyl acetate content:
11.2 %; saponification degree: 40 %; $X \times Y/100$: 4.48) being used
as an ethylene-vinyl acetate copolymer layer.

(Comparative Example 3)

Layer structure: the same as Example 1

The same as those used in Example 1 except that EVA
(ULTRACENE 540 available from TOSO CORPORATION; vinyl acetate
5 content: 3.5 %; saponification degree: 0 %; X x Y/100: 0) being
used as an ethylene-vinyl acetate copolymer layer.

(Comparative Example 4)

Layer structure: the same as Example 1

As a TFE copolymer, one prepared as follows was used:

10 The powder of the fluorine-containing polymer used in
Example 1 (190 kg) and pure water (200 liters) were charged in
a stainless steel tank. To the mixture, 28 % aqueous ammonia was
added and then the mixture was stirred at 80°C for 5 hours while
stirring. The powdery material was recovered from the tank,
15 washed with water and dried to obtain a powdery polymer.

Table 1

	TFE copolymer	Adhesive layer (Et-vinyl acetate copolymer layer)	Maximum adhesive strength (initial) (kg/cm)	Adhesive strength after dipping in fuel (kg/cm)	Dissolving test of adhesive layer in fuel
Ex. 1	(Ex. 1)	EVOH $X \times Y/100$ =64.6	Resin breakage >7.38	2.30	No dissolving
Ex. 2	(Ex. 1)	EVOH $X \times Y/100$ =11.2	2.82	1.78	No dissolving
Comp. Ex. 1	(Ex. 1)	(Epoxy- modified polyethy- lene)	Resin breakage >4.06	X Spontaneously peeled due to dissolving of adhesive layer	Dissolved
Ex. 3	(Ex. 1)	EVOH $X \times Y/100$ =9.52	2.64	X Spontaneously peeled due to dissolving of adhesive layer	Dissolved
Comp. Ex. 2	(Ex. 1)	EVOH $X \times Y/100$ =4.48	X Sponta- neously peeled		Dissolved
Comp. Ex. 3	(Ex. 1)	EVA $X \times Y/100$ =0	X Sponta- neously peeled		Dissolved
Comp. Ex. 4	(Ex. 1) (amide terminal)	EVOH $X \times Y/100$ =64.6	X Sponta- neously peeled		No dissolving

CLAIMS

1. A laminate comprising

(1) a layer of a tetrafluoroethylene copolymer comprising 30 to 81 % by mole of tetrafluoroethylene and 70 to 19 % by mole of at least one other monomer and having a carbonate group in a polymer chain or at a polymer chain terminal, which has a melt flow rate of 0.1 to 100 g/10 minutes (230°C, 5 kg-load) and a melting point of 90 to 230°C,

(2) a layer of an ethylene-vinyl acetate copolymer formed on one surface of the layer (1), which satisfies the following relationship:

$$X \times Y/100 \geq 7.0$$

wherein X is a vinyl acetate content (% by mole) and Y is a saponification degree of a methyl ester (%), and

(3) a layer of a polyolefin resin formed on the layer (2).

2. The laminate according to claim 1, wherein said tetrafluoroethylene copolymer of the layer (1) has a melt flow rate of 0.1 to 100 g/10 minutes (200°C, 5 kg-load) and a melting point of 90 to 200°C is preferably used.

3. The laminate according to claim 1 or 2, wherein said tetrafluoroethylene copolymer comprises 40 to 81 % by mole of tetrafluoroethylene and 60 to 19 % by mole of other monomer.

4. The laminate according to any one of claims 1 to 3, wherein said tetrafluoroethylene copolymer has a melt flow rate of 1.0 to 100 g/10 minutes.

5. The laminate according to any one of claims 1 to 3, wherein said tetrafluoroethylene copolymer has a melt flow rate of 1.0 to 50 g/10 minutes.

6. The laminate according to any one of claims 1 to 5, wherein said tetrafluoroethylene copolymer has a critical shear rate of 10 to 10^3 sec^{-1} at 230°C .

7. The laminate according to any one of claims 1 to 6, wherein
5 said tetrafluoroethylene copolymer comprises tetrafluoroethylene, ethylene, a fluorovinyl compound of the formula (I):



wherein Rf is a fluoroalkyl group having 2 to 10 carbon atoms,
10 and hexafluoropropylene, in which a molar ratio of tetrafluoroethylene to ethylene is from 40:60 to 90:10, the content of said fluorovinyl compound is from 0 to 10 % by mole (based on the whole copolymer) and the content of hexane is from 0 to 30 % by mole (based on the whole copolymer).

15 8. The laminate according to any one of claims 1 to 7, wherein said ethylene-vinyl acetate copolymer has a vinyl acetate content X (% by mole) and a saponification degree Y (%) of a methyl ester satisfy the following relationship:

$$X \times Y/100 \geq 10.0.$$

20 9. The laminate according to any one of claims 1 to 8, wherein said ethylene-vinyl acetate copolymer has a melt flow rate (200°C , 5 kg-load) of 0.5 to 100 g/10 minutes.

10. A tubular member comprising a laminate according to any one of claims 1 to 9.

25 11. The tubular member according to claim 10, which is a tube for fuels.

12. A container comprising a laminate according to any one of claims 1 to 9.

13. The container according to claim 12, which is a fuel tank.

14. A film comprising a laminate according to any one of claims 1 to 9.

5 15. A sheet comprising a laminate according to any one of claims 1 to 9.

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65
66
67
68
69
70
71
72
73
74
75
76
77
78
79
80
81
82
83
84
85
86
87
88
89
90
91
92
93
94
95
96
97
98
99
100
101
102
103
104
105
106
107
108
109
110
111
112
113
114
115
116
117
118
119
120
121
122
123
124
125
126
127
128
129
130
131
132
133
134
135
136
137
138
139
140
141
142
143
144
145
146
147
148
149
150
151
152
153
154
155
156
157
158
159
160
161
162
163
164
165
166
167
168
169
170
171
172
173
174
175
176
177
178
179
180
181
182
183
184
185
186
187
188
189
190
191
192
193
194
195
196
197
198
199
200
201
202
203
204
205
206
207
208
209
210
211
212
213
214
215
216
217
218
219
220
221
222
223
224
225
226
227
228
229
230
231
232
233
234
235
236
237
238
239
240
241
242
243
244
245
246
247
248
249
250
251
252
253
254
255
256
257
258
259
260
261
262
263
264
265
266
267
268
269
270
271
272
273
274
275
276
277
278
279
280
281
282
283
284
285
286
287
288
289
290
291
292
293
294
295
296
297
298
299
300
301
302
303
304
305
306
307
308
309
310
311
312
313
314
315
316
317
318
319
320
321
322
323
324
325
326
327
328
329
330
331
332
333
334
335
336
337
338
339
340
341
342
343
344
345
346
347
348
349
350
351
352
353
354
355
356
357
358
359
360
361
362
363
364
365
366
367
368
369
370
371
372
373
374
375
376
377
378
379
380
381
382
383
384
385
386
387
388
389
390
391
392
393
394
395
396
397
398
399
400
401
402
403
404
405
406
407
408
409
410
411
412
413
414
415
416
417
418
419
420
421
422
423
424
425
426
427
428
429
430
431
432
433
434
435
436
437
438
439
440
441
442
443
444
445
446
447
448
449
450
451
452
453
454
455
456
457
458
459
460
461
462
463
464
465
466
467
468
469
470
471
472
473
474
475
476
477
478
479
480
481
482
483
484
485
486
487
488
489
490
491
492
493
494
495
496
497
498
499
500
501
502
503
504
505
506
507
508
509
510
511
512
513
514
515
516
517
518
519
520
521
522
523
524
525
526
527
528
529
530
531
532
533
534
535
536
537
538
539
540
541
542
543
544
545
546
547
548
549
550
551
552
553
554
555
556
557
558
559
560
561
562
563
564
565
566
567
568
569
570
571
572
573
574
575
576
577
578
579
580
581
582
583
584
585
586
587
588
589
590
591
592
593
594
595
596
597
598
599
600
601
602
603
604
605
606
607
608
609
610
611
612
613
614
615
616
617
618
619
620
621
622
623
624
625
626
627
628
629
630
631
632
633
634
635
636
637
638
639
640
641
642
643
644
645
646
647
648
649
650
651
652
653
654
655
656
657
658
659
660
661
662
663
664
665
666
667
668
669
670
671
672
673
674
675
676
677
678
679
680
681
682
683
684
685
686
687
688
689
690
691
692
693
694
695
696
697
698
699
700
701
702
703
704
705
706
707
708
709
710
711
712
713
714
715
716
717
718
719
720
721
722
723
724
725
726
727
728
729
730
731
732
733
734
735
736
737
738
739
740
741
742
743
744
745
746
747
748
749
750
751
752
753
754
755
756
757
758
759
760
761
762
763
764
765
766
767
768
769
770
771
772
773
774
775
776
777
778
779
780
781
782
783
784
785
786
787
788
789
790
791
792
793
794
795
796
797
798
799
800
801
802
803
804
805
806
807
808
809
810
811
812
813
814
815
816
817
818
819
820
821
822
823
824
825
826
827
828
829
830
831
832
833
834
835
836
837
838
839
840
841
842
843
844
845
846
847
848
849
850
851
852
853
854
855
856
857
858
859
860
861
862
863
864
865
866
867
868
869
870
871
872
873
874
875
876
877
878
879
880
881
882
883
884
885
886
887
888
889
890
891
892
893
894
895
896
897
898
899
900
901
902
903
904
905
906
907
908
909
910
911
912
913
914
915
916
917
918
919
920
921
922
923
924
925
926
927
928
929
930
931
932
933
934
935
936
937
938
939
940
941
942
943
944
945
946
947
948
949
950
951
952
953
954
955
956
957
958
959
960
961
962
963
964
965
966
967
968
969
970
971
972
973
974
975
976
977
978
979
980
981
982
983
984
985
986
987
988
989
990
991
992
993
994
995
996
997
998
999
1000

ABSTRACT

A layer of a tetrafluoroethylene copolymer comprising 30 to 81 % by mole of tetrafluoroethylene and 70 to 19 % by mole of at least one other monomer and having a carbonate group in a polymer chain or at a polymer chain terminal, which has a melt flow rate of 0.1 to 100 g/10 minutes (230°C, 5 kg-load) and a melting point of 90 to 230°C and a layer of a polyolefin resin are adhered with a layer of an ethylene-vinyl acetate copolymer, which satisfies the following relationship: $X \times Y/100 \geq 7.0$ wherein X is a vinyl acetate content (% by mole) and Y is a saponification degree of a methyl ester (%). The resulting laminate has high resistance to fuels, and the layer of the tetrafluoroethylene copolymer having a carbonate group and the layer of the polyolefin resin are firmly adhered.

BIRCH, STEWART, KOLASCH & BIRCH, LLP

P.O. Box 747 • Falls Church, Virginia 22040-0747
Telephone: (703) 205-8000 • Facsimile: (703) 205-8050

PLEASE NOTE:
YOU MUST
COMPLETE THE
FOLLOWING

COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT AND DESIGN APPLICATIONS

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated next to my name; that I verily believe that I am the original, first and sole inventor (if only one inventor is named below) or an original, first and joint inventor (if plural inventors are named below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Insert Title:

LAMINATE COMPRISING FLUORINE-CONTAINING POLYMER

Fill in Appropriate
Information -
For Use Without
Specification
Attached:

the specification of which is attached hereto. If not attached hereto,
the specification was filed on _____ as
United States Application Number _____;
and amended on _____ (if applicable) and/or
the specification was filed on August 24, 2000 as PCT
International Application Number PCT/JP00/05673; and was
amended under PCT Article 19 on _____ (if applicable)

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I do not know and do not believe the same was ever known or used in the United States of America before my or our invention thereof, or patented or described in any printed publication in any country before my or our invention thereof or more than one year prior to this application, that the same was not in public use or on sale in the United States of America more than one year prior to this application, that the invention has not been patented or made the subject of an inventor's certificate issued before the date of this application in any country foreign to the United States of America on an application filed by me or my legal representative or assigns more than twelve months (six months for designs) prior to this application, and that no application for patent or inventor's certificate on this invention has been filed in any country foreign to the United States of America prior to this application by me or my legal representatives or assigns, except as follows.

I hereby claim foreign priority benefits under Title 35, United States Code, §119(a)-(d) of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

Priority Claimed

Insert Priority
Information:
(if appropriate)

<u>238227/1999</u> (Number)	<u>Japan</u> (Country)	<u>Aug. 25, 1999</u> (Month/Day/Year Filed)	<input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Month/Day/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Month/Day/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
_____ (Number)	_____ (Country)	_____ (Month/Day/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No

I hereby claim the benefit under Title 35, United States Code, §119(e) of any United States provisional application(s) listed below.

Insert Provisional
Application(s):
(if any)

_____ (Application Number)	_____ (Filing Date)
_____ (Application Number)	_____ (Filing Date)

All Foreign Applications, if any, for any Patent or Inventor's Certificate Filed More than 12 Months (6 Months for Designs) Prior to the Filing Date of This Application:

Insert Requested
Information:
(if appropriate)

Country	Application Number	Date of Filing (Month/Day/Year)
_____	_____	_____
_____	_____	_____

I hereby claim the benefit under Title 35, United States Code, §120 of any United States and/or PCT application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States and/or PCT application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose information which is material to the patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

Insert Prior U.S.
Application(s):
(if any)

_____ (Application Number)	_____ (Filing Date)	_____ (Status - patented, pending, abandoned)
_____ (Application Number)	_____ (Filing Date)	_____ (Status - patented, pending, abandoned)

I hereby appoint the following attorneys to prosecute this application and/or an international application based on this application and to transact all business in the Patent and Trademark Office connected therewith and in connection with the resulting patent based on instructions received from the entity who first sent the application papers to the attorneys identified below, unless the inventor(s) or assignee provides said attorneys with a written notice to the contrary:

Raymond C. Stewart
Joseph A. Kolasch
Bernard L. Sweeney
Charles Gorenstein
Leonard R. Svensson
Andrew D. Meikle
Joe McKinney Muncy
John W. Bailey
Gary D. Yacura

(Reg. No. 21,066)
(Reg. No. 22,463)
(Reg. No. 24,448)
(Reg. No. 29,271)
(Reg. No. 30,330)
(Reg. No. 32,868)
(Reg. No. 32,334)
(Reg. No. 32,881)
(Reg. No. 35,416)

Terrell C. Birch
James M. Slattery
Michael K. Mutter
Gerald M. Murphy, Jr.
Terry L. Clark
Marc S. Weiner
Donald J. Daley
John A. Castellano

(Reg. No. 19,382)
(Reg. No. 28,380)
(Reg. No. 29,680)
(Reg. No. 28,977)
(Reg. No. 32,644)
(Reg. No. 32,181)
(Reg. No. 34,313)
(Reg. No. 35,094)

Send Correspondence to:

BIRCH, STEWART, KOLASCH & BIRCH, LLP

or Customer No. 2292

P.O. Box 747 • Falls Church, Virginia 22040-0747

Telephone: (703) 205-8000 • Facsimile: (703) 205-8050

PLEASE NOTE:
YOU MUST
COMPLETE
THE
FOLLOWING:

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full Name of First or Sole Inventor: Insert Name of Inventor Insert Date This Document is Signed	GIVEN NAME/FAMILY NAME <u>Tatsuya HIGUCHI</u>	INVENTOR'S SIGNATURE <u>Tatsuya Higuchi</u>	DATE* <u>Feb. 19, 2002</u>
Insert Residence Insert Citizenship	Residence (City, State & Country) <u>Settsu-shi, Osaka Japan</u>	CITIZENSHIP <u>Japan</u>	
Insert Post Office Address	POST OFFICE ADDRESS (Complete Street Address including City, State & Country) <u>c/o Yodogawa Works of DAIKIN INDUSTRIES, LTD., 1-1, Nishihitotsuya, Settsu-shi, Osaka 566-8585 Japan</u>		
Full Name of Second Inventor, if any: see above	GIVEN NAME/FAMILY NAME <u>Hidenori OZAKI</u>	INVENTOR'S SIGNATURE <u>Hidenori Ozaki</u>	DATE* <u>Feb. 19, 2002</u>
	Residence (City, State & Country) <u>Settsu-shi, Osaka Japan</u>	CITIZENSHIP <u>Japan</u>	
	POST OFFICE ADDRESS (Complete Street Address including City, State & Country) <u>c/o Yodogawa Works of DAIKIN INDUSTRIES, LTD., 1-1, Nishihitotsuya, Settsu-shi, Osaka 566-8585 Japan</u>		
Full Name of Third Inventor, if any: see above	GIVEN NAME/FAMILY NAME <u>Takahiro KITAHARA</u>	INVENTOR'S SIGNATURE <u>Takahiro Kitahara</u>	DATE* <u>Feb. 19, 2002</u>
	Residence (City, State & Country) <u>Settsu-shi, Osaka Japan</u>	CITIZENSHIP <u>Japan</u>	
	POST OFFICE ADDRESS (Complete Street Address including City, State & Country) <u>c/o Yodogawa Works of DAIKIN INDUSTRIES, LTD., 1-1, Nishihitotsuya, Settsu-shi, Osaka 566-8585 Japan</u>		
Full Name of Fourth Inventor, if any: see above	GIVEN NAME/FAMILY NAME <u>Kazuo ISHIWARI</u>	INVENTOR'S SIGNATURE <u>Kazuo Ishiwari</u>	DATE* <u>Feb. 19, 2002</u>
	Residence (City, State & Country) <u>Settsu-shi, Osaka Japan</u>	CITIZENSHIP <u>Japan</u>	
	POST OFFICE ADDRESS (Complete Street Address including City, State & Country) <u>c/o Yodogawa Works of DAIKIN INDUSTRIES, LTD., 1-1, Nishihitotsuya, Settsu-shi, Osaka 566-8585 Japan</u>		
Full Name of Fifth Inventor, if any: see above	GIVEN NAME/FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
	Residence (City, State & Country)	CITIZENSHIP	
	POST OFFICE ADDRESS (Complete Street Address including City, State & Country)		